values at the higher energies, they are larger than the corresponding Bethe-Heitler cross section.

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Small Oscillation Theory of the Interaction of a Particle and Scalar Field

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We study the interaction of a nonrelativistic particle with a scalar field, with particular application to the theory of polarons. The approach is based on a general classical method for the integration of equations of motion. The Hamiltonian is transformed by successive canonical transformations, the first corresponding to describing the motion relative to special solutions of the equations of motion. This stage as applied to suitably ordered Heisenberg equations of motion is identical with intermediate coupling theory. The second transformation treats the coupled small oscillations of particle and field oscillators about the chosen special solution. This affords a natural extension of intermediate coupling theory for this problem. Differences between the classical and quantum theories arise in the ordering of operators; the differences play a crucial role in determining the effective cutoff in wave vector space.

1. INTRODUCTION

THE theory of the interaction of an electron with lattice vibrations has received much attention recently. Aside from the question of oversimplifications customarily made in describing the physical system, there is the problem of finding quantum theoretic methods powerful enough to analyze the structure of the typical Hamiltonians encountered. The present deficiency hampers progress in the theory of normal and super conducting metals at low temperatures, and makes uncertain, estimates of lattice mobility in polar crystals and semiconductors. The electron-lattice interaction is also interesting from the point of view of the theory of elementary particles. The Hamiltonian corresponds to a simplified nonrelativistic field theory with a *bona fide* cutoff. One is concerned with developing theories of source recoil for intermediate and strong coupling which may have implications for the general theory of fields. The studies have already given rise to a new adiabatic theory.[1]

We will here be chiefly concerned with the polaron problem—loosely speaking, the theory of the interaction of a free electron with the optical modes of a polar lattice. It possesses the simplification that a minimum energy $\hbar\omega$ is required to excite a quantum of optical vibration (ω is the common frequency of the vibrations). For the lowest states of the system, one has a pure self energy situation with no free quanta present. A range of such states is possible; they are characterized by an integral of the motion $\mathbf{P}$ with the dimensions of a momentum, where $\mathbf{P}$ satisfies $P^2/2m \lesssim \hbar\omega$. Most studies[2–5] have dealt with the properties of the above class of states as a function of coupling strength, in particular the energy of the very lowest state with $P=0$, and the nearby states for which $P^2/2m \ll \hbar\omega$. Less attention has been paid to the case where $P^2/2m \approx \hbar\omega$ and to the nature of the excited states. These regions are of interest in the theory of dielectric breakdown and in mobility calculations. The determination of the energies of the low-lying states has involved mainly variational techniques. The most successful methods are those of Hohler and Feynman. The advantages are known; the disadvantages are that it is hard to make a picture of the behavior, that the intrinsic structure of the Hamiltonian is lost sight of and that the determination of the excited states is made difficult.

In the following, we note that classical theory points to a natural attack on the problem, and we explore some of the consequences. In the theory of orbits one finds a solution of the equations of motion (stable orbit) for which the initial conditions have been chosen in a particular way. Then one studies small oscillations about the orbit. If the orbit is stable, there is a region of phase space in which the full freedom in choice of initial conditions is present. At the edges of the region of stable motion the small-oscillation assumption is not valid; however, classically the amplitude may be made arbitrarily small so that there is always a domain where anharmonic terms can be neglected. For the

[1] N. Bogoliubov, Ukr. Math. J. T II, No. 2, 3 (1950); S. Tyablikov, Zhur. Eksptl. i Teort. Fiz. 21, 377 (1951).

[2] S. I. Pekar, *Untersuchungen uber die Elektronentheorie der Kristalle* (Akademische-Verlag, Berlin, 1954).
[3] H. Fröhlich, Advances in Phys. 3, 325 (1954).
[4] G. Hohler, Z. Physik 140, 192 (1955).
[5] R. P. Feynman, Phys. Rev. 97, 660 (1955). These papers contain references to earlier work.

polaron problem there is such a stable motion; the electron moves with uniform momentum and the lattice modes undergo forced oscillations. There are then neighboring states in which electron and lattice oscillators undergo coupled small oscillation. This classical picture will be applied here to the lowest states; the intimately phased motion of electrons and ions leads to lowering of the energy, and may be of the type needed in the theory of superconductivity.

Mathematically both classical and quantum theory involve a series of canonical transformations.[6] The first introduces coordinates relative to the stable orbit, and the second is a normal mode transformation; later transformations are perturbation treatments of the anharmonic terms. However, because of the finite de Broglie wavelength of the electron and the zero-point motion of the lattice, the classical and quantum transformations differ. The differences are most significant for short-wavelength modes and provide a cutoff in the quantum theory not present classically. This advantage is offset by the fact that the concept of small oscillations is not entirely natural in quantum theory, since the minimum amplitude of zero-point vibrations may take a problem inherently anharmonic. This is reflected in the mathematical formalism in questions of ordering of creation and annihilation operators, and in approximations beyond the classical scheme.

2. CLASSICAL THEORY

For details of notation and the analysis leading to the Hamiltonian we refer to Fröhlich *et al.*[7] The Hamiltonian is

$$H=p^2/2m+\tfrac{1}{2}\sum_k\{(Y_k^2/M)+M\omega_k^2X_k^2\}+\sum g_k'\{X_k\sin(\mathbf{k}\cdot\mathbf{q})+(Y_k/M\omega_k)\cos(\mathbf{k}\cdot\mathbf{q})\},\quad(1)$$

where $\mathbf{p}$ and $\mathbf{q}$ are canonical coordinates of the particle of mass m; ω_k is the frequency of a lattice oscillator with wave vector $\mathbf{k}$; Y_k and X_k are canonical momenta and coordinates of the oscillators; M is a constant with dimensions of the square of a time; X_k^2 and Y_k^2/M have dimensions of energy; g_k' has dimensions of a square root of energy. We may write $g_k'=g_k/L^{\frac{3}{2}}$, where L is the length of the cube enclosing the system (periodic boundary conditions). Then $\mathbf{k}$ takes on the values $\mathbf{k}=\mathbf{n}2\pi/L$, where the triplet $\mathbf{n}$ consists of negative and positive integers.

One verifies that the following is a special solution of Hamilton's equations for the system[8]:

$$\mathbf{p}=\mathbf{p}_0=\text{const};\qquad \mathbf{q}=(\mathbf{p}_0/m)t+\mathbf{q}_0;$$
$$X_k=\frac{g_k'}{M\omega_k}\frac{\sin(\mathbf{k}\cdot\mathbf{q})}{(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k};\quad Y_k=g_k'\frac{\cos(\mathbf{k}\cdot\mathbf{q})}{(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k}.\quad(2)$$

[6] The approach is a special case of the general method for integration of classical equations of motion. See Whittaker, *Analytical Dynamics* (Dover Publications, New York, 1944), Chap. 16.

[7] Fröhlich, Pelzer and Zienau (hereafter denoted F.P.Z.), Phil. Mag. **41**, 221 (1950).

[8] E. P. Gross, Technical Report No. 55, Laboratory for Insulation Research, Massachusetts Institute of Technology, December, 1952 (unpublished).

Since there are no terms with a time dependence at the frequency of the free oscillations of the lattice, the solution must be closely related to the quantum solution for the lowest state where no free quanta are present. The quantum eigenstate will be seen however to involve at least an additional range of classical states, in which particles and oscillators undergo coupled forced oscillations of a particular type in the vicinity of the special classical solution.

The general equations have, in addition to the energy, the integral of the motion

$$\mathbf{P}=\mathbf{p}+\sum_{\mathbf{k}}\frac{\mathbf{k}}{\omega_k}\left(\frac{Y_k^2}{2M}+\frac{M\omega_k^2}{2}X_k^2\right).\quad(3)$$

The special solution (2) yields for H and $\mathbf{P}$;

$$\mathbf{P}=\mathbf{p}_0+\sum_{\mathbf{k}}\frac{\mathbf{k}}{2M\omega_k}\frac{g_k'^2}{[(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k]^2},$$
$$H=\frac{p_0^2}{2m}+\sum\frac{g_k'^2/M\omega_k}{(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k}+\tfrac{1}{2}\sum\frac{g_k'^2/M}{[(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k]^2}.\quad(4)$$

The special solution thus involves a relationship between H and $\mathbf{P}$ through the intermediary of $\mathbf{p}_0$.

Equations (2) and (4) have singularities for waves whose phase velocity $\omega\mathbf{k}/k^2$ is at the particle velocity $\mathbf{p}_0/m$. If one restricts consideration to sufficiently small values of $\mathbf{p}_0$ so that $|\mathbf{k}\cdot\mathbf{p}_0/m|<|\omega_k|$ for all oscillators, the results depend on the cut-off value of k. For a classical electron in a crystal the maximum value of k is given by the lattice spacing, and the singularity means that excitation of natural lattice oscillations must be considered. Quantum theory provides a cutoff arising from wavelength of the particle. For weak coupling this has been discussed by F.P.Z. and is $k_{\max}\sim(2m\omega/\hbar)^{\frac{1}{2}}$ which leads to an energy proportional to g^2. One can also estimate the effective mass for small $\mathbf{p}_0$ by using (4). For strong coupling the cutoff will depend on the strength of interaction, representing a tendency to confine the particle in a potential well. The question of the behavior of the cutoff is thus vital, and is inherently quantum mechanical.

We now go further into the structure of the equations of motion by studying small oscillations. The results are simplified by making the following canonical transformation:

$$\mathbf{P}=\mathbf{p}+\sum_{\mathbf{k}}\frac{\mathbf{k}}{\omega_k}\left(\frac{Y_k^2}{2M}+\frac{M\omega_k^2X_k^2}{2}\right),\quad\mathbf{Q}=\mathbf{q},\quad(5)$$
$$Q_k'=X_k\cos(\mathbf{k}\cdot\mathbf{q})-\frac{Y_k}{M\omega_k}\sin(\mathbf{k}\cdot\mathbf{q}),$$
$$\frac{P_k'}{M\omega_k}=X_k\sin(\mathbf{k}\cdot\mathbf{q})+\frac{Y_k}{M\omega_k}\cos(\mathbf{k}\cdot\mathbf{q}).$$

In terms of the new variables the Hamiltonian is

$$H=\frac{P^2}{2m}+\sum g_k'\frac{P_k'}{M\omega_k}+\frac{1}{2M}\sum_{\mathbf{k}}\left(1-\frac{\mathbf{k}\cdot\mathbf{P}}{m\omega_k}\right)\times(P_k'^2+M^2\omega_k^2Q_k'^2)+\frac{1}{(2M)^2}\cdot\frac{1}{2m}\times\left\{\sum\frac{\mathbf{k}}{\omega_k}(P_k'^2+M^2\omega_k^2Q_k'^2)\right\}^2. \quad (6)$$

The solution (2) corresponds now to a simple shift in the new momentum coordinates,

$$Q_k'=Q_k^0=0, \quad P_k'=P_k^0=\frac{g_k'}{(\mathbf{k}\cdot\mathbf{p}_0/m)-\omega_k}, \quad (7)$$

and

$$\mathbf{P}=\mathbf{p}_0+\textstyle\sum_{\mathbf{k}}(\mathbf{k}P_k^{0\,2}/2M\omega_k).$$

To introduce variables describing the motion relative the stable orbit, we perform the following canonical transformation:

$$P_k=P_k'-P_k^0, \quad Q_k=Q_k'. \quad (8)$$

Then $H=\sum_{i=0}^{4}H_i$, with $H_1=0$ and

$$H_0=\frac{p_0^2}{2m}+\sum_k g_k'\frac{P_k^0}{M\omega_k}+\frac{M}{2}\sum_k\omega_k^2\left(\frac{P_k^0}{M\omega_k}\right)^2,$$
$$H_2=\frac{1}{2m}\left(\sum_{\mathbf{k}}\frac{\mathbf{k}P_k^0P_k}{M\omega_k}\right)^2+\frac{1}{2M}\sum_{\mathbf{k}}\left(1-\frac{\mathbf{k}\cdot\mathbf{p}_0}{m\omega_k}\right)\times(P_k^2+M^2\omega_k^2Q_k^2). \quad (9)$$

We have not written down the cubic and quartic parts of H. The constant part H_0 is just the energy of Eq. (4). The next stage in the solution is to find the normal modes for H_2. This is always possible in principle, but here H_2 has a particularly simple form which permits explicit calculation of the normal frequencies and modes. We write

$$Q_k=\textstyle\sum_\lambda R_{k\lambda}\xi_\lambda, \quad P_k=\sum_\lambda R_{k\lambda}\eta_\lambda, \quad (10)$$

where ξ_λ and η_λ are the new canonical coordinates and momenta and $R_{k\lambda}$ is a real, orthogonal matrix. The quadratic portion of the Hamiltonian is diagonal in terms of the new coordinates with Ω_λ as the normal frequencies. The $R_{k\lambda}$ and Ω_λ are determined by writing the equations of motion using only H_2, and inserting into them Eqs. (10) together with

$$\ddot{\eta}_\lambda=-\Omega_\lambda^2\eta_\lambda.$$

We find

$$(\rho_k^2-\Omega_\lambda^2)R_{k\lambda}=-\frac{\rho_k\mathbf{k}P_k^0}{m_k}\cdot\sum_{\mathbf{l}}\frac{\mathbf{l}P_l^0R_{l\lambda}}{M\omega_l}, \quad (11)$$

where $\rho_k=\omega_k(1-\mathbf{k}\cdot\mathbf{p}_0/m\omega_k)$.

There are two types of solutions. In the first type,

$$\beta_\lambda\equiv\sum_{\mathbf{l}}\frac{\mathbf{l}P_l^0R_{l\lambda}}{M\omega_l}=0.$$

For a given λ $R_{k\lambda}=0$ unless $\rho_k^2=\Omega_\lambda^2$. The Ω_λ are simply the original lattice frequencies, (as modified by the Doppler shift), relative to a particle moving with velocity $\mathbf{p}_0/m$. Only a small fraction of the lattice oscillators satisfy $\rho_k^2=\Omega_\lambda^2$, and these are modes built out of a small number of oscillators combined so as to form standing waves with a node at the position of the electron. This is the reason why the frequencies are unmodified by interaction.

The second type has $\beta_\lambda\neq0$ and

$$R_{k\lambda}=-\frac{\rho_kP_k^0}{m}\frac{\mathbf{k}\cdot\beta_\lambda}{(\rho_k^2-\Omega_\lambda^2)}. \quad (12)$$

The β_λ are determined by the normalization condition on $R_{k\lambda}$. The Ω_λ are determined from

$$\beta_\lambda=-\sum_{\mathbf{k}}\frac{\rho_kP_k^{0\,2}\mathbf{k}(\mathbf{k}\cdot\beta_\lambda)}{mM\omega_k(\rho_k^2-\Omega_\lambda^2)}. \quad (13)$$

These modes are built up from small contributions from all the lattice oscillators and contain the coupling constant for the electron lattice interaction. The normal frequencies are found between the poles of the right-hand side of (13).

3. QUANTUM TREATMENT

Because of the similarity of the operator equations of motion in the Heisenberg representation and the classical equations the classical theory may be taken over. The new points all involve the appropriate ordering of noncommutative factors. We introduce the creation and destruction operators

$$\left\{\begin{matrix}b_k^*\\ b_k\end{matrix}\right\}=\left(\frac{M\omega_k}{2\hbar}\right)^{\frac{1}{2}}(Q_k'\mp iP_k'/M\omega_k), \quad (14)$$

satisfying $[b_k,b_l^*]=\delta_{kl}$. Introduce the purely imaginary quantity (dimensions of energy),

$$V_k=-ig_k'(\hbar/2M\omega_k)^{\frac{1}{2}}, \quad (15)$$

and rewrite Eq. (6) as

$$H=\frac{P^2}{2m}+\sum\hbar\left(\omega_k-\frac{\mathbf{k}\cdot\mathbf{P}}{m}+\frac{\hbar^2k^2}{2m}\right)b_k^*b_k+\sum(V_kb_k+V_k^*b_k^*)+\sum_{\mathbf{k},\mathbf{l}}\frac{\hbar^2}{2m}(\mathbf{k}\cdot\mathbf{l})b_k^*b_l^*b_kb_l. \quad (16)$$

We have made use of the fact that the zero-point

momentum $\sum\hbar\mathbf{k}/2$ is zero and have discarded the zero-point energy $\sum\hbar\omega_k/2$. The operators have been ordered so that the creation operators stand to the left of the destruction operators; this arrangement brings out the term $\sum(\hbar^2k^2/2m)b_k^*b_k$ from the quartic part of the Hamiltonian (6). This term comes originally from the $p^2/2m$ term, i.e., from the particle kinetic energy and represents physically the DeBroglie wavelength cutoff. Now perform the shift, Eq. (8).[9] The ordering of operators is unaltered. We have

$$\begin{aligned} a_k&=b_k+f(k), \quad a_k^*=b_k^*+f^*(k),\\ Q_k&=Q_k', \quad P_k=P_k'-P_k^0. \end{aligned} \tag{17}$$

Here $a_k=(M\omega_k/2\hbar)^{\frac{1}{2}}\{Q_k+iP_k/M\omega_k\}$, and we will later show that $f(k)$ is purely imaginary. Thus $P_k^0=-(2M\hbar\omega_k)^{\frac{1}{2}}if(k)$. We will discuss the choice of $f(k)$ in detail below. Equations (9) are replaced by $H=\sum_{n=0}^{4}\times H^{(n)}$, where

$$\begin{aligned} H^{(0)}&=\frac{P^2(1-\eta)^2}{2m}+\sum_{\mathbf{k}}\left(\hbar\omega_k+\frac{\hbar^2k^2}{2m}\right)|f(k)|^2,\\ H^{(1)}&=\sum\{\hbar\rho_kf(\mathbf{k})+V_k^*\}a_k^*+\text{c.c.},\\ H^{(2)}&=\sum\hbar\rho_ka_k^*a_k+(\hbar/2m)\sum_{\mathbf{k},\mathbf{l}}\mathbf{k}\cdot\mathbf{l}f(k)f(l)\{a_k^*a_l^*\\ &\qquad -a_k^*a_l+a_ka_l-a_l^*a_k\},\\ H^{(3)}&=(\hbar^2/m)\{\sum\mathbf{k}a_k^*f(k)\cdot\sum\mathbf{l}a_l^*a_l\\ &\qquad +\sum\mathbf{l}a_l^*a_l\cdot\sum\mathbf{k}a_kf^*(k)\},\\ H^{(4)}&=(\hbar^2/2m)\sum\mathbf{k}\cdot\mathbf{l}a_k^*a_l^*a_ka_l, \end{aligned} \tag{18}$$

where now

$$\begin{aligned} \rho_k&=\omega_k-\frac{\mathbf{k}\cdot\mathbf{P}}{m}(1-\eta)+\frac{\hbar^2k^2}{2m},\\ \eta\mathbf{P}&=\sum\mathbf{k}|f(\mathbf{k})|^2. \end{aligned} \tag{19}$$

One can now choose $f(\mathbf{k})$ so that $H^{(1)}$ is zero. This ensures that there are no constant "forces" in the Heisenberg equations of motion of the operators a_k and a_k^*. Thus there are no linear increases of these operators with time and we have found the stable orbit. As in the classical case, there are contributions from the quartic terms of the original equation (6) to the determination of $f(\mathbf{k})$. A reason for adopting the above order is that the terms $H^{(2)}$, $H^{(3)}$, $H^{(4)}$ have no first-order contribution to the energy of the ground state in a perturbation calculation in a representation where $a_k^*a_k$ is diagonal. The results then agree with quantum perturbation theory in the limit of weak coupling.

This stage of the procedure is precisely the intermediate coupling theory of the polaron.[9–11] Here we have been motivated by a study of the equations of motion of the operators rather than by a variational approach. The fact that the diagonal matrix elements of $H^{(2)}$, $H^{(3)}$, $H^{(4)}$ are zero means that the true ground-state energy is below the value obtained with H_0.

Now our object is to take into account the coupled vibrations of electron and lattice oscillators. We shall therefore find the normal modes of $H^{(2)}$. If one takes f as in the intermediate coupling theory, we find that including effects of the oscillations gives a solution valid for larger coupling strengths and lowers the energy at all coupling strengths. However, one does not obtain results valid in the strong coupling limit. This is connected with an important point. The a_k will be linear combinations of new creation and annihilation operators belonging to the new normal modes. Thus, the Hamiltonian will not be appropriately ordered in terms of the new variables. In so ordering, $H^{(3)}$ contains linear terms and the process of finding the normal modes destroys the stable orbit. It is therefore more appropriate to keep f free until the normal modes have been found and to determine it by then setting the linear terms equal to zero.

Now introduce coordinates and momenta appropriate to the Doppler-shifted waves. We have

$$\begin{Bmatrix} a_k\\ a_k^* \end{Bmatrix}=\left(\frac{M\rho_k}{2\hbar}\right)^{\frac{1}{2}}(G_k-iF_k/M\rho_k). \tag{20}$$

Then

$$\begin{aligned} G_k&=Q_k(\omega_k/\rho_k)^{\frac{1}{2}}, \quad F_k=P_k(\rho_k/\omega_k)^{\frac{1}{2}},\\ G_k&=\textstyle\sum_sT_{ks}\xi_s, \quad F_k=\sum_sT_{ks}\eta_s. \end{aligned}$$

$H^{(2)}$ becomes

$$H^{(2)}=\sum\left(\frac{F_k^2}{2M}+\frac{M\rho_k^2}{2}G_k^2\right)-\frac{1}{M\hbar m}\left(\sum\frac{\hbar\mathbf{k}fF_k}{\rho_k^{\frac{1}{2}}}\right)^2. \tag{21}$$

The lowest states are characterized by zero quanta in each of the normal modes. There will therefore be a zero-point energy $\sum_s(\hbar\Omega_s/2)$. Thus, the contribution of $H^{(2)}$ to the energy is

$$\Delta E^{(2)}=\sum_s\frac{\hbar\Omega_s}{2}-\sum_k\frac{\hbar\rho_k}{2}-\frac{\hbar^2}{2m}\sum_k k^2|f(k)|^2. \tag{22}$$

The reordering of $H^{(4)}$ gives in general an additional contribution to the energy which can be found after the normal modes are determined.

4. ENERGY OF LOWEST STATE

In this section, we treat the lowest state of the polaron for which $\mathbf{P}=0$. For simplicity of presentation we go through the calculations for a one-dimensional case. Measure energies in units of $\hbar\omega$, momenta in units of $(2m\hbar\omega)^{\frac{1}{2}}$, and introduce

$$t_k=(\rho_k/\omega)^2; \quad s=(\Omega_s/\omega)^2.$$

[9] Lee, Low, and Pines, Phys. Rev. **90**, 297 (1953).
[10] M. Gurari, Phil. Mag. **44**, 329 (1953).
[11] S. Tyablikov, Zhur. Eksptl. i Teort. Fiz. **25**, 688 (1953).

Consider modes of type 1, and label them with the index s_0. Then the frequencies are given by

$$s_0(k)=t_k=(1+k^2)^2. \tag{23}$$

For a given $s_0>0$, there are two values of k:

$$k_\pm(s_0)=\pm[(s_0)^{\frac{1}{2}}-1]^{\frac{1}{2}}$$

Since one must satisfy $\sum (kfT_{ks0}/\rho_k^{\frac{1}{2}})=0$, and since $f(k)$ is an even function of k for $P=0$, we have

$$T_{k+s0}=T_{k-s0}=1/\sqrt{2}. \tag{24}$$

The last step follows from the normalization condition

$$T_{k+s0}{}^2+T_{k-s0}{}^2=1.$$

Thus, for normal modes of type (1) and $P=0$,

$$\xi_{s0}=(1/\sqrt{2})(Q_{k+}+Q_{k-}). \tag{25}$$

There is no shift in zero-point energy for type-1 modes.

The frequencies of type-2 modes are determined from

$$-1=\sum_k \frac{4(t_k)^{\frac{1}{2}}k^2f^2(k)}{(s-t_k)}, \tag{26}$$

with $t_k=(1+k^2)^2$ or $k^2=t_k^{\frac{1}{2}}-1$. This type of eigenvalue equation has been treated in the literature.[12,13] The poles of the right-hand side are at $s=t_k$ with $k=2\pi n/L$ the spacing of poles is

$$\epsilon(t_n)=t_{n+1}-t_n=(2n_{+1})(2\pi/L)^2\{(2\pi/L)^2 \times(2n+1)+2+2(2\pi n/L)^2\}.$$

Inserting the value of $2\pi n/L$ as a function of t_n, we have as $L\to\infty$:

$$\epsilon(t_n)\to 4(2\pi/L)(t_n{}^{\frac{1}{2}}-1)^{\frac{1}{2}}t_n{}^{\frac{1}{2}}, \tag{27}$$

which gives the spacing as a function of frequency.

Let s_m be the root lying between t_m and t_{m+1}. Then introduce the shift σ_m

$$s_m=\{1+(2\pi m/L)^2\}^2+\sigma_m, \quad 0\leq\sigma_m<\epsilon(t_m). \tag{28}$$

The ratio σ_m/ϵ_m is less than unity and tends to a non-zero value as $L\to\infty$, but $\sigma_m\to 0$ as $L\to\infty$. The total shift is zero-point energy of type-2 modes is therefore (in dimensionless units),

$$\tfrac{1}{2}\sum_{\text{all } s_m}(s_m{}^{\frac{1}{2}}-t_m{}^{\frac{1}{2}})=\tfrac{1}{4}\sum_{s_m}(\sigma_m/t_m{}^{\frac{1}{2}}), \tag{29}$$

where we keep only the first term in the expansion of $s_m{}^{\frac{1}{2}}$ since $\sigma_m\to 0$ as $L\to\infty$ and t_m is finite. The energy shift is then

$$\tfrac{1}{4}\sum_{sm}\frac{\sigma_m(s_m)}{\epsilon_m(s_m)}\frac{\epsilon_m(s_m)}{\sqrt{t_m}}\to\tfrac{1}{4}\int_{s=1}^{\infty}\frac{\sigma(s)}{\epsilon(s)}\frac{ds}{\sqrt{s}}, \tag{30}$$

since $\Delta s\to\epsilon_m$ as $L\to\infty$. To find the ratio $\sigma(s)/\epsilon(s)$, one uses the result

$$\sum_{n=-\infty}^{+\infty}\frac{\alpha_n{}^2}{s-t_n}=\mathrm{P}\int\frac{\alpha^2(t)dt}{s-t}+\pi\alpha^2(s)\cot\left(\frac{\pi\sigma(s)}{\epsilon(s)}\right), \tag{31}$$

where P stands for the principal value of the integral and

$$\alpha^2(t_n)=\alpha_n{}^2/\epsilon(t_n). \tag{32}$$

For our problem, $\alpha_n{}^2=4t_n{}^{\frac{1}{2}}(n2\pi/L)^2f^2(2\pi n/L)$, so that

$$\alpha^2(t)=(L/2\pi)f^2[(t^{\frac{1}{2}}-1)^{\frac{1}{2}}][(t^{\frac{1}{2}}-1)]^{\frac{1}{2}}. \tag{33}$$

Inserting Eq. (31) into the eigenvalue equation, we find

$$\frac{\pi\sigma(s)}{\epsilon(s)}=\tan^{-1}\left\{-\pi\alpha^2(s)\Big/\left(1+\mathrm{P}\int_1^\infty\frac{\alpha^2(t)dt}{s-t}\right)\right\}. \tag{34}$$

The normal modes of type 2 are related to the original lattice coordinates by the transformation coefficients:

$$T_{ks}{}^{(2)}=-\frac{4t_k{}^{\frac{1}{2}}f(k)\beta_sk}{t_k-s}; \quad \beta_s=\sum_k\frac{kf(k)T_{ks}{}^{(2)}}{\sqrt{\rho_k}}. \tag{35}$$

The $T_{ks}{}^{(2)}$ satisfy the orthogonality relation:

$$\textstyle\sum_k T_{ks}{}^{(2)}T_{ks'}{}^{(2)}=\delta_{ss'}, \tag{36}$$

as may be seen by breaking the left-hand side of (36) into partial fractions and using the eigenvalue equation (26). The β_s are normalization coefficients and are determined by

$$\frac{1}{|\beta_s|^2}=\sum_k\frac{4t_k|\alpha_k|^2}{(t_k-s)^2}. \tag{37}$$

To evaluate this we need the sum[13] $\sum_n[\gamma_n{}^2/(t_n-s)^2]$, where $\gamma^2(t)=\gamma_n{}^2/\epsilon_n(t)=4t|\alpha(t)|^2$. In the limit $L\to\infty$,

$$|\beta_s|^2\to\frac{2\pi}{L}\frac{(s^{\frac{1}{2}}-1)^{\frac{1}{2}}}{\pi^2(s)^{\frac{1}{2}}|\alpha^2(s)|}\sin^2(\pi\sigma/\epsilon). \tag{38}$$

For type-two modes, we therefore have

$$\xi_s=\textstyle\sum_k T_{sk}{}^{(2)}Q_k. \tag{39}$$

The inverse transformation giving the Q_k and P_k in terms of the ξ_s, η_s will be needed in order to rewrite the Hamiltonian in terms of the new variables. A given Q_k involves both types of normal modes. We have

$$Q_k=(1/\sqrt{2})\xi_{s0(k)}+\textstyle\sum_s T_{ks}{}^{(2)}\xi_s, \tag{40}$$

where s_0 is the single value of s for the type one mode arising from k.

The above information concerning the transformation coefficients enables us to complete the calculation of the energy. We introduce creation and annihilation

[12] Wigner, Critchfield, and Teller, Phys. Rev. **56**, 530 (1939).
[13] N. V. Kampen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 15 (1951).

operators for the normal modes:

$$\begin{Bmatrix} d_s \\ d_s^* \end{Bmatrix} = \left(\frac{M\Omega_s}{2\hbar}\right)^{\frac{1}{2}} (\xi_s \pm i\eta_s/M\Omega_s), \tag{41}$$

and

$$\xi_s = (\hbar/2M\Omega_s)^{\frac{1}{2}}\{d_s + d_s^*\},$$
$$i\eta_s/M\Omega_s = (\hbar/2M\Omega_s)^{\frac{1}{2}}\{d_s - d_s^*\}.$$

We work in a representation where $d_s^* d_s$ is diagonal and consider the ground state Ψ_0 for which $d_s\Psi_0 = 0$. Then $H^{(1)}$ and $H^{(3)}$ have an odd number of creation and annihilation operators and thus have the property

$$\langle\Psi_0, H^{(1)}\Psi_0\rangle = \langle\Psi_0, H^{(3)}\Psi_0\rangle = 0.$$

The energy of the system by our method is therefore

$$E = \langle\Psi_0, H\Psi_0\rangle = H^{(0)} + \langle\Psi_0, H^{(2)}\Psi_0\rangle + \langle\Psi_0, H^{(4)}\Psi_0\rangle. \tag{42}$$

The true energy must always be less than this value.

We now show that the quartic contribution to the energy is zero (for $P=0$). One has

$$\langle\Psi_0, H^{(4)}\Psi_0\rangle = (\hbar/2m)\sum_{k,l}(\mathbf{k}\cdot\mathbf{l})\langle a_l a_k\Psi_0, a_l a_k\Psi_0\rangle.$$

Now

$$a_k = \frac{d_{s_0(k)}}{\sqrt{2}} + \left(\frac{M\rho_k}{2\hbar}\right)^{\frac{1}{2}} \sum_s \frac{T_{ks}^{(2)}}{2}\left(\frac{\rho_k}{\Omega_s}\right)^{\frac{1}{2}} \times\left\{d_s\left(1+\frac{\Omega_s}{\rho_k}\right) + d_s^*\left(1-\frac{\Omega_s}{\rho_k}\right)\right\}. \tag{43}$$

We have made use of the fact that $s_0(k) = t_k$ (type-1 modes). Then

$$a_k\Psi_0 = \sum_s T_{ks}^{(2)}(\rho_k/\Omega_s)^{\frac{1}{2}}(1-\Omega_s/\rho_k)d_s^*\Psi_0.$$

The term $a_l a_k\Psi_0$ involves $T_{ks}T_{ls'}$ as a factor, and the entire inner product $\langle a_l a_k\Psi_0, a_l a_k\Psi_0\rangle$ involves $T_{ks}T_{k\mu} \times T_{ls'}T_{l\mu'}$ which is an even function of both k and l. The summation over k and l involves the odd factor $k\cdot l$ so that $\langle\Psi_0, H^{(4)}\Psi_0\rangle = 0$.

The complete expression for the energy is therefore $H^{(0)} + \langle\Psi_0, H^{(2)}\Psi_0\rangle$, where $f(k)$ can still be chosen freely. From this point of view, the method contains a variational principle. It will be of interest later, however, to seek to determine $f(k)$ in a natural way in accordance with our basic physical picture. The total energy is

$$E = \sum_k (V_k f + V_k^* f^*) + \sum_k \rho_k |f(k)|^2 - \sum k^2 |f|^2 + \frac{1}{4\pi}\int_{s=1}^{\infty} \frac{ds}{\sqrt{s}} \times\tan^{-1}\left\{-\pi\alpha^2(s) \Big/ \left(1 + \mathrm{P}\int_1^{\infty} \frac{\alpha^2(t)dt}{s-t}\right)\right\}. \tag{44}$$

In the one-dimensional problem, $V_k = V$ (a constant). Introduce the pure number

$$U^2 = [V^2/(\hbar\omega)^2]L(\hbar/2m\omega)^{-\frac{1}{2}}. \tag{45}$$

A simple choice for $f(k)$ is

$$f(k) = \frac{V}{1+k^2/b^2}. \tag{46}$$

For weak and intermediate coupling $b=1$, but for stronger coupling it depends on the interaction strength, i.e., the DeBroglie cutoff for a free particle is no longer appropriate. We have

$$\alpha^2(t) = \frac{U^2b^4}{2\pi}\frac{(t^{\frac{1}{2}}-1)^{\frac{1}{2}}}{(b^2+(t)^{\frac{1}{2}}-1)^2}, \tag{47}$$

$$\mathrm{P}\int_1^{\infty} \frac{\alpha^2(t)dt}{s-t} = \frac{U^2b^4}{2\pi s}\{K(s,b)+J(s,b)\}, \tag{48}$$

where

$$K(s,b) = \int_1^{\infty} \frac{(x-1)^{\frac{1}{2}}xdx}{(s+x)(x+b^2-1)^2} = -J(-s,b) + 2\pi \cdot\frac{2s(1+s)^{\frac{1}{2}}}{(b^2-s)^2} = \frac{2\pi}{b}\frac{2b(1+s)^{\frac{1}{2}}+1+b^2}{[b^2+(1+s)^{\frac{1}{2}}]^2},$$
$$J(s,b) = \mathrm{P}\int_1^{\infty} \frac{(x-1)^{\frac{1}{2}}xdx}{(s-x)(x+b^2-1)^2} = \frac{2\pi}{b} \times\frac{s(1-3b^2)-(b^2-1)^2}{(s-1+b^2)^2}. \tag{49}$$

The integrals have been evaluated by the method of residues. Examine first the case $b=1$ (intermediate coupling theory). Then the total energy of the system is

$$E = -\tfrac{3}{4}|U|^2 + \frac{1}{2\pi}\int_{r=1}^{\infty} \times\tan^{-1}\left\{\frac{(r-1)^{\frac{1}{2}}}{(r^2/|U|^2) - [(1+r)^{\frac{1}{2}}-2]}\right\}dr. \tag{50}$$

Now $\tan^{-1}x$ has the expansion

$$\tan^{-1}x = x - x^3/3 + x^5/5 \quad \text{for} \quad x^2 < 1.$$

For this expansion to be suitable for all r, we must have

$$|U|^2 < \min\frac{r^2}{|(r-1)^{\frac{1}{2}} - 2 + (1+r)^{\frac{1}{2}}|} \lesssim 1.6.$$

In addition, for

$$|U|^2 < \min\frac{r^2}{|(1+r)^{\frac{1}{2}} - 2|} \leq 1.6,$$

one can expand the denominator in powers of $|U|^2$. The result is

$$E = -\tfrac{1}{2}|U|^2 - |U|^4(1/6\pi - 1/16) + \cdots. \tag{51}$$

This result is probably accurate for $|U|^2$ somewhat

>1.6 since the region of r where convergence fails is small. The additional energy is always lower than $-\frac{1}{2}|U|^2$, representing an improvement over the intermediate coupling theory. However, for very large coupling strengths the term containing the $\tan^{-1}$ saturates, and gives a dependence which is slower than $|U|^2$. The energy is then roughly $-\frac{3}{4}|U|^2$ rather than the weak coupling value of $-\frac{1}{2}|U|^2$.

Thus with the choice of f suggested by intermediate coupling theory we do not get the correct strong coupling energy given by the adiabatic approximation ($E\sim-0.1|U|^4$). From the present point of view this arises because the intermediate coupling choice of f has implicit a cutoff related to the DeBroglie wavelength of the free particle. The cutoff in reality must depend on the coupling strength, so as to represent the "binding" of the particle in the potential well created when it polarizes the lattice.

We have checked numerically that letting $b^2=1+\varphi(U)$ improves the energy in the region $|U|^2\approx10$, but have not discovered whether b^2 can be chosen as a function of coupling strength so as to obtain the strong coupling limit. In the next section, a 'natural' way to specify f is discussed.

The energy for the three-dimensional situation is easily expressed in terms of the one-dimensional results. The eigenvalue equation is a vector relation:

$$\beta_s=4\sum_k \frac{(t_k)^{\frac{1}{2}}f^2(\mathbf{k})}{(t_k-s)}\mathbf{k}(\mathbf{k}\cdot\beta_s). \tag{52}$$

There are three independent modes of type 2 for a given frequency s. We take these to have β_s along the x, y, z axes respectively. To find the frequency shift of these modes, consider, for example, the mode with β_s along the z axis. Then

$$-1=4\sum_k \frac{(t_k)^{\frac{1}{2}}k_z^2f^2(\mathbf{k})}{s-t_k}. \tag{53}$$

For the $P=0$ case $f(\mathbf{k})$ and t_k depend only on the absolute value of k. We may therefore perform the angle integrations and find

$$1\to4\cdot\frac{4\pi}{3}\left(\frac{L}{2\pi}\right)^3\int_0^\infty\left\{\frac{k^2(t_k)^{\frac{1}{2}}f^2(k)}{t_k-s}\right\}k^2dk. \tag{54}$$

The eigenvalue equation is of the same form as the one-dimensional one. Here we take

$$\alpha_n{}^2=\frac{8\pi}{3}\left(\frac{2\pi n}{L}\right)^4t_n{}^{\frac{1}{2}}f^2\left(\frac{2\pi n}{L}\right)\left(\frac{L}{2\pi}\right)^2 \tag{55}$$

or

$$\alpha^2(t)=(t^{\frac{1}{2}}-1)^{\frac{3}{2}}f^2[(t^{\frac{1}{2}}-1)^{\frac{1}{2}}]\left(\frac{L}{2\pi}\right)^3\cdot\tfrac{2}{3}\pi.$$

With this expression for $\alpha^2(t)$, the energy shift of the type-2 modes is then 3 times the value given by Eq. (35). Corresponding to the one-dimensional choice (46), in three dimensions we may take

$$f(k)=V_k/(1+k^2/b^2).$$

In dimensionless units,[7,9]

$$V_k=-(i/k)(4\pi\alpha'/L^3)^{\frac{1}{2}}.$$

Insert in Eq. (55) for $\alpha^2(t)$ and compare with Eq. (33) for one dimension. With $|U|^2=\frac{2}{3}\alpha'$ and with the factor 3, the arc-tangent term in Eq. (44) is correct for the three-dimensional case. The total energy is then

$$E=\sum(V_kf+V_k{}^*f^*)+\sum|f(\mathbf{k})|^2+\frac{3}{4\pi}\int_{s=1}^\infty\frac{ds}{\sqrt{s}}\times\tan^{-1}\left\{-\pi\alpha^2(s)\Big/\left(1+\mathrm{P}\int_1^\infty\frac{\alpha^2(t)dt}{s-t}\right)\right\}. \tag{56}$$

Again, as in one dimension, the three independent type-1 normal modes of a given frequency are standing waves with modes at the particle.

5. REORDERING OF $H^{(3)}$

We return to the question of the systematic determination of f. The diagonalization of $H^{(2)}$ destroys the ordering of the cubic term. We investigate the process of determining f by the requirement that the linear terms in the Hamiltonian vanish when it is expressed in terms of ordered operators for the new normal modes. We order $H^{(3)}$, as given in Eq. (19), so that the d_s stand to the right of the $d_s{}^*$ and select the linear portion.

Examine first $\sum kfa_k$ (for $P=0$). From (43), noting that $d_{s0(k)}=d_{s0(-k)}$ one sees that there is no contribution from type-1 modes. Then for the one-dimensional case,

$$\sum kfa_k=\sum_{k,s}kf(k)\frac{T_{ks}{}^{(2)}}{\sqrt{2}}\left(\frac{\rho_k}{\Omega_s}\right)^{\frac{1}{2}}\times\left\{d_s\left(1+\frac{\Omega_s}{\rho_k}\right)+d_s{}^*\left(1-\frac{\Omega_s}{\rho_k}\right)\right\}. \tag{57}$$

Next consider $\sum ka_k{}^*a_k$. Since $T_{ks}{}^{(2)}$ is an odd function of k, only the terms consisting of products of one operator of type 1 and one of type 2 will survive the summation over k.

$$\sum ka_k{}^*a_k=\sum_{k,s}\frac{kT_{ks}{}^{(2)}}{2}\left(\frac{\rho_k}{\Omega_s}\right)^{\frac{1}{2}}\left[\left\{d_s\left(1+\frac{\Omega_s}{\rho_k}\right)+d_s{}^*\left(1-\frac{\Omega_s}{\rho_k}\right)\right\}\frac{d_{s0(k)}{}^*}{\sqrt{2}}+\text{c.c.}\right]. \tag{58}$$

The linear portion of $H^{(3)}$ is then [using $f^*(k)$

$=-f(k)]$,

$$H_{\text{lin}}^{(3)}=\frac{\hbar^2}{m}\sum_{k,l,s}(\mathbf{k}\cdot\mathbf{l})f(l)\frac{Tl_s^{(2)}T_{ls}^{(2)}}{\sqrt{2}}\left(\frac{\rho_l}{\rho_k}\right)^{\frac{1}{2}}\times(1-\Omega_s/\rho_l)\{d_{s0(k)}-d_{s0(k)}^*\}. \quad (59)$$

Since $d_{s0(k)}=d_{s0(-k)}$ and $d_{s0(k)}=(1/\sqrt{2})(a_k+a_{-k})$, one can express $H_{\text{lin}}^{(3)}$ in terms of the a_k, a_k^*.

$$H_{\text{lin}}^{(3)}=\frac{\hbar^2}{m}\sum_{k,l,s}(\mathbf{k}\cdot\mathbf{l})f(l)T_{ks}^{(2)}T_{ls}^{(2)}\left(\frac{\rho_l}{\rho_k}\right)^{\frac{1}{2}}\times\left(1-\frac{\Omega_s}{\rho_k}\right)\{a_k-a_k^*\}. \quad (60)$$

Combining this with $H^{(1)}$, we determine $f(k)$ by setting the coefficients of a_k, a_k^* equal to zero. The condition is

$$\rho_k f^*(k)+V_k+k\sum_{l,s}lf(l)T_{ls}^{(2)}T_{ks}^{(2)}\left(\frac{\rho_l}{\rho_k}\right)^{\frac{1}{2}}\left(1-\frac{\Omega_s}{\rho_l}\right)=0.$$

Now insert expressions (35) and (36). We find

$$f(k)=V_k\Big/\left(1+4k^2\sum_s\frac{\Omega_s\rho_k\beta_s^2}{(\rho_k^2-\Omega_s^2)}\right).$$

The $\sum_s$ term is positive since $\beta_s^2<0$, and the root Ω_s is larger than the corresponding root for zero coupling. The effect of ordering of the cubic term is thus a modification of the cutoff of $f(k)$, in a way that depends on coupling strength; the change is more complicated than that of Eq. (46).

With the help of Eqs. (27), (31), (35), and (38), one finds

$$4\sum_s\frac{\Omega_s\rho_k\beta_s^2}{\rho_k^2-\Omega_s^2}=\frac{1+\mathrm{P}\int\frac{\alpha^2(t)dt}{\rho_k^2-t}}{(\pi\alpha^2)^2+\left(1+\mathrm{P}\int\frac{\alpha^2dt}{\rho_k^2t}\right)^2}+\mathrm{P}\int\frac{\rho_k ds\,\sin^2[\pi\sigma(s)/\epsilon]}{\pi^2s^{\frac{1}{2}}\alpha^2(s)(\rho_k^2-s)}. \quad (62)$$

As the coupling strength $\to 0$ the sum tends to 1. In principle $f(k)$ can be obtained from this nonlinear integral equation with a degenerate kernel. By direct variation of the energy of Eq. (56), it can be shown that f satisfying Eq. (61) yields the lowest energy attainable by the present method. We have not succeeded in using the results of this section to find the proper explicit form of $f(k)$ for large coupling strengths.

CONCLUSIONS

Our results underscore the value of analysis of the structure of the classical equations of motion. If this can be done by canonical methods, so that a portion of the transformed Hamiltonian describes a set of classical states accurately, there is the basis for an attack on the properties of some quantum eigenstates. In the present problem one is led to a natural extension of intermediate coupling theory. It is interesting that the quantum modifications arising from the ordering of operators affect mainly the proper cutoff. The first step, leading to intermediate coupling theory, replaces a classical theory sensitive to the maximum value of k by a quantum theory with a cutoff arising from the electron wavelength; the effect comes from ordering of nonlinear terms in the equations of motion. The second step of treating the quadratic terms modifies the cutoff as discussed in Sec. 5. The approach is readily applied to the states $P=0$; this will be done elsewhere.

The important question whether the best $f(k)$ yields correct results for strong coupling has not been resolved.[14] If not, one would like to know whether anharmonic classical motions contribute, or whether classically forbidden 'motions' are needed. One should keep in mind that $H^{(4)}$ may contain quadratic terms, if reordered in terms of creation and annihilation operators for the normal modes. It appears formidable to simultaneously remove linear terms, and have quadratic terms diagonal and anharmonic terms "properly" ordered; this may be an unwarranted extension of the small-oscillation concept to the quantum theory.

[14] *Note added in proof.*—Investigations of T. Schultz show that results inferior to the adiabatic approximation are obtained in the limit of strong coupling.